

Infrared Hall conductivity of $\text{Na}_{0.7}\text{CoO}_2$

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We report infrared Hall conductivity $\sigma_{xy}(\omega)$ of $\text{Na}_{0.7}\text{CoO}_2$ thin films determined from Faraday rotation angle θ_F measurements. $\sigma_{xy}(\omega)$ exhibits two types of hole conduction, Drude and incoherent carriers. The coherent Drude carrier shows a large renormalized mass and Fermi liquid-like behavior of Hall scattering rate, $\gamma_H \sim aT^2$. The spectral weight is suppressed and disappears at $T = 120\text{K}$. The incoherent carrier response is centered at mid-IR frequency and shifts to lower energy with increasing T . Infrared Hall constant is positive and almost independent of temperature in sharp contrast with the dc-Hall constant.

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The sodium cobalt oxide Na_xCoO_2 has attracted much attention recently since the discovery of large thermopower at $x > 0.7$ ¹ and $T=5\text{K}$ superconductivity at $x=0.3$.² The material has a layered crystal structure in which edge-sharing CoO_6 octahedra form 2D plane separated by the charge-providing Na^{+2} spacer. The CoO_2 plane has triangular bonding symmetry which brings about geometrical frustration of the Co spins. Carrier transport in this conducting plane can potentially lead to exotic phase such Anderson's resonant valence bond (RVB) state or non-s wave superconducting pairing.³ With varying Na content, x , Na_xCoO_2 exhibits a rich phase diagram which includes the superconductivity ($x=0.35$),² charge ordered insulating state ($x=0.5$),⁴ Curie-Weiss metal ($x=0.7$), and spin-density wave state ($x>0.65$).⁵ The large thermopower observed at high x also makes the material interesting for applications.¹

Na_xCoO_2 with $x=0.7$ is a host compound of the series and it shows its own unusual properties at low temperature. Dc resistivity shows linear- T behavior at $T \leq 100\text{K}$. The quasiparticles seen in angle resolved photoemission (ARPES) experiment decreases in intensity with T and disappears at $T \geq 100\text{K}$.⁶ Also, in spite of moderate metallicity ($\rho = 0.1\text{m}\Omega$ at 30K), the magnetic susceptibility data is not Pauli-like but rather the Co spins show local magnetic moments. The Hall coefficient increases with temperature without saturation up to as high as $T=500\text{K}$.⁷ The positive Hall sign changes to negative at $T < 200\text{K}$. These anomalous properties suggest that Na_xCoO_2 is another example of a strongly correlated electron system.

In $\text{Na}_{0.7}\text{CoO}_2$, the active Co t_{2g} bands are split by crystal field into e_g and d_{z^2} bands. The e_g band is fully occupied. The electronically active d_{z^2} band is half filled (occupied by 1 electron per Co) at $x=0$ and adding electrons at $x>0$ can lead to a doped Mott-Hubbard system where the fundamental physics is exotic and not well understood and therefore it is currently subject of intense study.⁸

In this paper, we report infrared Hall effect of $\text{Na}_{0.7}\text{CoO}_2$ from Faraday angle measurements on thin film samples. The experiment probes the Hall con-

ductivity σ_{xy} at an infrared frequency. The study of σ_{xy} provides invaluable informations on charge dynamics of strongly correlated metal, often complementary to the conventional optical conductivity σ_{xx} as successfully demonstrated in high- T_c superconductor YBaCuO ,^{9,10} $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$,¹¹ and electron-doped $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$.¹²

Epitaxial $\gamma\text{-Na}_{0.7}\text{CoO}_2$ thin films (1000\AA thick) were grown by pulsed laser deposition technique on SrTiO_3 substrates. The films have (0001) orientation where the CoO_2 hexagonal plane is parallel with the substrate plane. Details of the sample growth and characterization was published elsewhere.¹³ We measured dc-resistance and dc-Hall effect of the films for $4\text{K} \leq T \leq 350\text{K}$ range. Both results showed similar temperature dependent results as those for single crystal $\text{Na}_{0.7}\text{CoO}_2$, suggesting high quality of the films. Faraday rotation angle of the film was measured using polarized infrared CO_2 laser with frequency $\omega = 1100\text{ cm}^{-1}$ and a photoelastic modulator at $30\text{K} \leq T \leq 300\text{K}$. The substrate contribution was independently measured and subtracted from data. The details of measurement technique are described elsewhere.¹⁴

Figure 1 shows the infrared complex Faraday angle θ_F (real and imaginary part) of a $\text{Na}_{0.7}\text{CoO}_2$ film at $\omega = 1100\text{ cm}^{-1}$. The data was reproducible in 8 traces on separate days. As T decreases from 300K , $\text{Re}(\theta_F)$ (and $\text{Im}(\theta_F)$) increases (decreases) gradually. At $T \leq 100\text{K}$, $\text{Re}(\theta_F)$ decreases with lowering T opposite to its high- T behavior and with a larger slope. The $\text{Im}(\theta_F)$ exhibits a sudden step-like jump at $T=100\text{K}$. This sudden change in θ_F represents, as we will discuss, the onset of contributions from coherent quasiparticles below 100K .

The transport functions of interest, the Hall angle, $\sigma_{xy}(\omega)$, and the Hall coefficient are derived from the Faraday angle data together with the optical conductivity $\sigma_{xx}(\omega)$, the film thickness, and substrate index from the Fresnel analysis of the thin film magneto optics as has been described elsewhere¹⁴. $\sigma_{xx}(\omega)$ of single crystal Na_xCoO_2 has been reported in the literature from reflectivity or ellipsometry measurements for different x 's.^{7,15,16,17} To analyze the magneto optical data, we

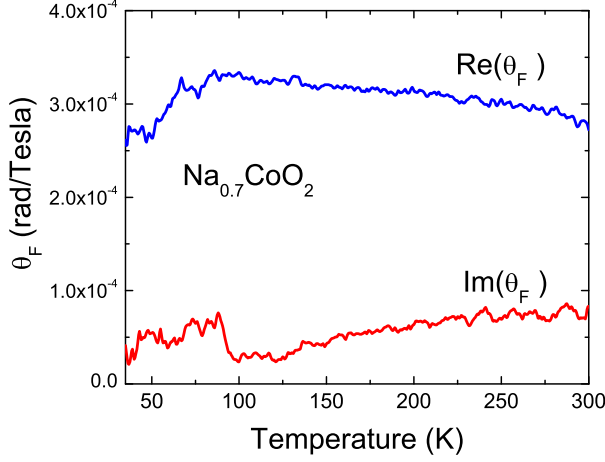


FIG. 1: Complex Faraday rotation angle θ_F of $\text{Na}_{0.7}\text{CoO}_2$ film measured at infrared frequency $\omega = 1100 \text{ cm}^{-1}$.

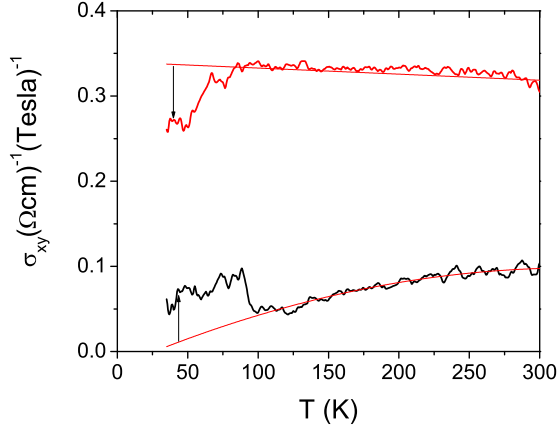


FIG. 2: Infrared Hall conductivity σ_{xy} . Dashed lines show polynomial fits of the high temperature part of data. The arrows represent the low-T deviations from the fit.

employ the $\sigma_{xx}(\omega)$ of $x=0.7^7$ and also nearby compositions $x=0.75^{15}$ and $x=0.82^{16}$. The $\sigma_{xx}(\omega)$ spectrum of $\text{Na}_{0.75}\text{CoO}_2$ shows that the optical conductivity consists of a Drude peak in the Far-IR range, plus a broad incoherent absorption at mid-infrared frequencies. These two components show up differently in $\sigma_{xy}(\omega)$. Near the frequency of our measurement, $\sigma_{xx}(\omega)$ is only weakly temperature dependent.

We will focus on the Hall conductivity $\sigma_{xy}(\omega)$ because it is the most fundamental magneto-transport response function. Fig. 2 shows $\sigma_{xy}(\omega)$ derived from θ_F and $\sigma_{xx}(\omega)$. $\text{Re}(\sigma_{xy})$ and $\text{Im}(\sigma_{xy})$ show similar T-dependence as θ_F because $\sigma_{xx}(\omega)$ is practically featureless and depends only weakly on temperature. To understand the

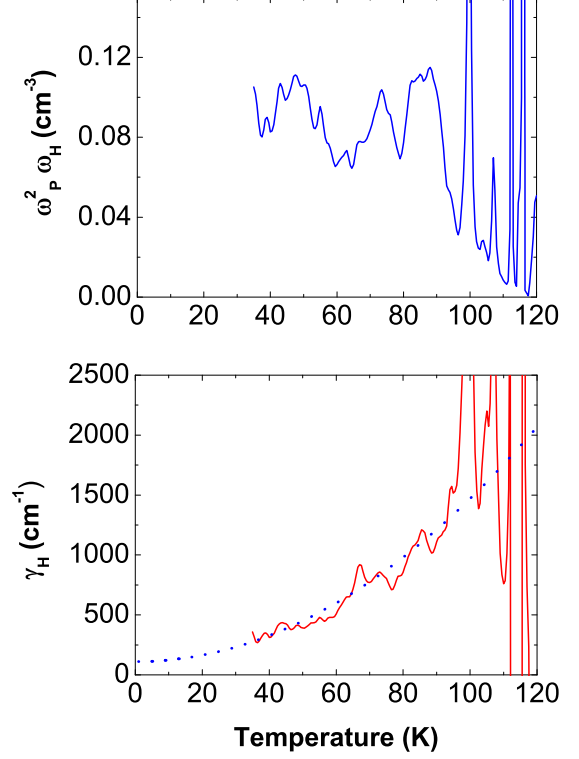


FIG. 3: Drude carrier analysis of the low-T $\Delta\sigma_{xy}$. (a) Squared plasma frequency $\omega_p^2(T)$ times Hall frequency $\omega_H(T)$ (b) Hall scattering rate $\gamma_H(T)$. Dashed line shows a fit with $\gamma_H(T) = \gamma_0 + aT^2$.

interesting low-T behavior, we first fit the high-T data with a simple polynomial in T curve shown as dashed lines, and assume that they extend to $T=0$. We label the deviations of σ_{xy} from the fit as $\Delta\sigma_{xy}(\omega) = \text{Re}(\Delta\sigma_{xy}) + i \cdot \text{Im}(\Delta\sigma_{xy})$. We attribute $\Delta\sigma_{xy}(\omega)$ to the coherent response of quasiparticles in $\text{Na}_{0.7}\text{CoO}_2$ which we model using a Drude conductivity form,

$$\Delta\sigma_{xy}(\omega) = \frac{\omega_p^2 \omega_H}{4\pi} \cdot \frac{1}{(\gamma_H - i\omega)^2} \quad (1)$$

Here ω_p and ω_H are the plasma frequency and the Hall frequency, respectively. γ_H is the Hall scattering rate. The former quantities are written as $\omega_p^2 = 4\pi \frac{nq^2}{m^*}$ and $\omega_H = \frac{qB}{m_H c}$ where n is the carrier density, m^* and m_H are the effective and Hall mass, respectively. We used Eq.(2) to extract $\omega_p^2 \omega_H$ and γ_H as a function of temperature from the data. These results are displayed in Fig.3.

$\omega_p^2 \omega_H(T)$ is seen to decrease as T increases and vanishes by $T \sim 120 \text{ K}$. Positive ω_H corresponds to hole conduction, $q > 0$. $\gamma_H(T)$ is observed to increase continuously with T. Close to 120 K, the data become increasingly noisy because $\Delta\sigma_{xy}$ is small in this T-range, and

it enters into denominator in calculating ω_p^2 and $\gamma_H(T)$. Angle resolved photoemission spectroscopy (ARPES) of $\text{Na}_{0.7}\text{CoO}_2$ by Hasan *et al*⁶ showed that Fermi surface consists of a large hole pocket centered at Γ point of Brillouin zone. The Fermi surface is nearly isotropic with $k_F=0.65\text{\AA}^{-1}$. The Fermi surface volume corresponds to carrier density $n\cong 8.4\cdot 10^{21}\text{ cm}^{-3}$ or 0.32 hole per Co plane. It is comparable to the nominal hole doping $1-x=0.3$. Using this n in $\omega_p^2 = \frac{4\pi e^3 B}{c} \frac{n}{m^* m_H}$ at $T=30\text{K}$, and assuming $m^* = m_H$ we find $m^*=9.3$. The large m^* shows strong renormalization of quasiparticle band, as seen by a weak dispersion of quasiparticle band in the ARPES data which also is reflected in electronic specific heat measurements.^{18,19} From n , m^* , and γ , we estimate dc resistivity $\rho = \frac{m^* \gamma}{ne^2} = 1.7 \cdot 10^{-4}\text{ }\Omega\text{ cm}$. This value is comparable with measured dc-value, 0.1 m $\Omega\text{ cm}$. If the suppression were due to a divergence of the effective mass $m_H \rightarrow \infty$ as $T \rightarrow 120\text{K}$, the electronic specific heat would show a diverging behavior at this temperature in contrast to the reported experimental data.²⁰ It is therefore more likely that the observed behavior comes from a reduction of coherent carrier density. The suppression of the quasiparticle peak was also seen in ARPES measurement.⁶

We observe that $\gamma_H(T)$ increases with T approximately as T^2 similar to a Fermi liquid. However, the magnitude of the scattering is large, $\gamma_H \gg T$ in contrast to the cuprates. Also the strong temperature dependence in the mid IR is in contrast with the observed behavior of the scattering rate deduced from $\sigma_{xx}(\omega)$ in $\text{Na}_{0.65}\text{CoO}_2$.¹⁵ At frequencies around 1000 cm^{-1} they find γ_{xx} to be independent of temperature for $T < 100\text{ K}$ and only weakly temperature dependent for $T > 100\text{ K}$. However, in their analysis they are assuming a one component model of conduction which we believe our IR Hall data refutes. The strong temperature dependence of γ_H in the mid IR is also not the expected behavior for a Fermi liquid. In Fermi liquids, the effective scattering rate for $\sigma_{xx}(\omega)$ is generally observed to behave as $\gamma \propto (\omega^2 + (\pi kT)^2)$. At 120K, the first term is 13 times the second term at our frequencies so that only a weak T -dependence would be expected.. This implies a weak ω dependence of γ_H in $\sigma_{xy}(\omega)$. Such anomalous behavior of γ_H has also been observed the optimally doped cuprates by IR Hall measurement and interpreted by Kotani.¹¹ In this theory, the suppression of the frequency dependence of γ_H is a consequence of current vertex corrections $\sigma_{xy}(\omega)$ for systems close to a spin density wave antiferromagnetic transition. $\text{Na}_{0.7}\text{CoO}_2$ is known to undergo a SDW transition at $T=20\text{K}$.⁵

The high- T part of $\sigma_{xy}(\omega)$ is associated with incoherent carriers. Hwang *et al* found that¹⁵ $\sigma_{xx}(\omega)$ consists of the Drude component and a broad mid-infrared incoherent peak. At low T , the latter has a localized line shape centered at $\sim 1000\text{ cm}^{-1}$. If we model it with a Lorentzian oscillator, its contribution to Hall conductivity

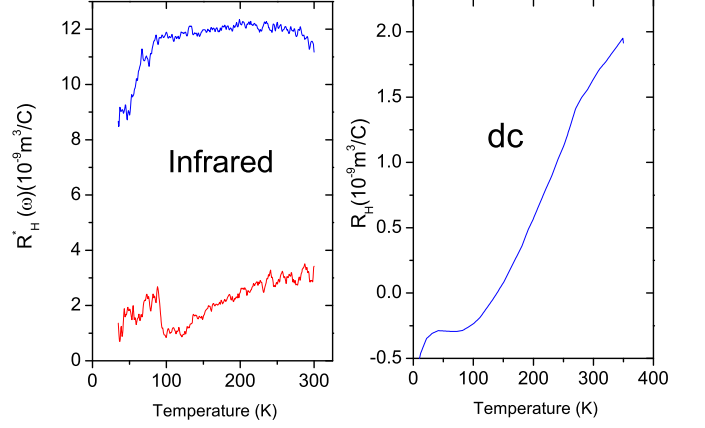


FIG. 4: Infrared Hall coefficient $R_H^*(\omega)$ and dc-Hall coefficient R_H .

ity $\sigma_{xy}(\omega)$ can be written as

$$\sigma_{xy}(\omega) = \frac{\Omega_p^2 \Omega_H}{4\pi} \cdot \frac{\omega^2}{(\omega^2 - \Omega_0^2 + i\omega\Gamma)^2} \quad (2)$$

where Ω_p^2 , Ω_0 , and Γ are the strength, center frequency, and width of the oscillator. Ω_H represent the the Hall frequency of the incoherent carriers. From $\sigma_{xx}(\omega)$ data, we adopt $\Omega_0=1000\text{ cm}^{-1}$, $\Gamma=5000\text{ cm}^{-1}$ and Ω_p^2 similar to that of the Drude ω_p^2 . Since Ω_0 almost coincides with ω ($=1100\text{ cm}^{-1}$) of our experiment, we have $\text{Im}(\sigma_{xy}) \cong 0$. This is consistent with our data extrapolated at $T=30\text{K}$. The positive σ_{xy} shows hole character and suggests that some of the doped holes become incoherent due to some localization process such as electron-phonon or electron-spin interaction. Such localization may explain the Curie-Weiss type magnetic susceptibility coexisting with the metallicity in $\text{Na}_{0.7}\text{CoO}_2$.²¹ In our model, Ω_0 is the most interesting parameter. If Ω_0 is assumed to decrease from 1000 cm^{-1} , $\text{Re}(\sigma_{xy})$ decreases and $\text{Im}(\sigma_{xy})$ increases, as the Hall conductivity data behaves with increasing T . $\sigma_{xy}(\omega)$ at $T=300\text{K}$ are produced if we take $\Omega_0 = 100\text{ cm}^{-1}$. This suggests that the incoherent peak shifts to lower frequency with temperature. The opposite T -dependences of $\text{Re}(\sigma_{xy})$ and $\text{Im}(\sigma_{xy})$, increasing and decreasing respectively with T , are produced only by the Ω_0 shift but not by other parameters of Eq.(2). In $\sigma_{xx}(\omega)$ the spectral weight is transferred from Drude part to higher frequency, $\omega < 1000\text{ cm}^{-1}$ with increasing T .¹⁵ The incoherent peak as a result gains spectral weight. Also the peak center effectively appears to shift to lower ω . At $T=300\text{K}$, $\sigma_{xx}(\omega)$ shows a localized line shape centered at FIR range 200 cm^{-1} .

The Dc-Hall effect of $\text{Na}_{0.7}\text{CoO}_2$ is also anomalous: The Hall coefficient R_H is negative at low- T . It becomes positive at $T > 200\text{K}$ and then increases linearly with T

without saturation up to as high as $T=500\text{K}$.⁴ In a non-interacting model Holstein showed that the Hall mobility on a triangular lattice is anomalously enhanced.²² Shastri *et al.* have studied an interacting case in the tJ model and found that the high frequency limit Hall coefficient $R_H^*(\omega)$ has a T-linear behavior similar to the measured DC- R_H .^{23,24} However, how this high frequency result relates to the DC R_H is not yet established. It is within this context, that we have studied the ac-Hall effect and compare it with the dc-Hall result. In Fig.4 we show the infrared Hall coefficient R_H^* obtained from $R_H^*(\omega) = \frac{\sigma_{xy}(\omega)}{\sigma_{xx}^2(\omega)}$ and compare it with the measured dc- R_H taken on the same film.

Note that the real part of $R_H^*(\omega)$ is nearly independent of T for $T > 100\text{K}$ in contrast to the dc result. Also there is no sign change. The imaginary part is much weaker than the real part. In strongly correlated electron systems, Shastri and Shraiman showed that the ac- $R_H^*(\omega)$ simplifies to the familiar semiclassical relation $R_H^* = \frac{1}{nqc}$, (n and q being the carrier density and charge).^{24,25} The relation is considered to become valid when the frequency is much higher than the carrier hopping energy t , ($\omega \gg t$) which applies to our case, $\omega = 1100\text{cm}^{-1}$, $t=11\text{ meV}$.⁶ The $R_H^*(\omega)$ then appears to show the constant density of hole carrier at $T > 100\text{K}$. The Hall constant at low frequency and dc-limit can be dominated by the self energy due to strong electron correlation. The non-simple behavior of dc- R_H may represents such effect. Clearly the infrared Hall effect provides a good starting reference to understand the anomalous behavior

of dc- R_H . At $T < 100\text{K}$, $\sigma_{xy}(\omega)$ have contributions from the quasiparticles and also the incoherent carriers so that $R_H^*(\omega)$ will be rather complicated.

In a heavy fermion(HF) metal such as CePd_3 and UPt_3 , the conduction carrier is hybridized with the local magnetic moment and leads a narrow low-frequency Drude conductivity. As T increases, the coherence is disturbed by thermal fluctuation. Then the Drude weight decreases and disappears at the Kondo temperature T_K .²⁶ In $\text{Na}_{0.7}\text{CoO}_2$, the holes coexist with the local Co magnetic moment of the Curie-Weiss like susceptibility. The effective mass m^* ($\cong 10$) is fairly large. Thus it is tempting to consider the Drude peak and the ω_p^2 suppression to stem from a similar origin. However, $T=100\text{K}$ is an order higher than typical HF which suggests a strong hybridization, while $m^*=10$, which is small in the HF standard, is inconsistent with that. Further study is needed to understand the unusual behavior of the Drude conductivity.

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